

Remarks

Claims 10 - 18 and 20, 21 are pending. Favorable reconsideration is respectfully requested.

The restriction is acknowledged but traversed, and claim 19 has been cancelled. Applicants respectfully disagree with the basis for the restriction, as there is a special technical feature common to the emulsion claims and process claim. However, since claim 14 contains all the limitations of claim 10, this process claim is subject to mandatory rejoinder upon a finding of allowable subject matter of claim 10.

Claims 15 - 18 have been objected to. Applicants respectfully traverse this rejection. A dependent claim must further limit the claim from which it depends. That is the case here. Claim 10 is directed to O/W and W/O emulsions, with no limitation as to their use. Claims 15 - 18 require the emulsion to be present in a coating material, adhesive, sealant, cleaning or cleansing agent, water repellent, adhesion promoter, release agent, paper coating, foam control agent, W/O/W, or O/WO multiple emulsion. All of these are further limiting, and all require the emulsion of claim 10 to be used. These are proper dependent claims and the objection is therefore respectfully traversed. However, to expedite prosecution, the claims have been amended to recite "formulated from" an emulsion of claim 10. It is noted that these claims correspond to "use claims" in the original priority application, which are allowable in both Germany and the European Union, but are prohibited in the U.S., and must be rewritten as either product claims or process claims.

Claims 10 - 13 and 15 - 18 have been rejected under 35 U.S.C. § 112 ¶2. Applicants respectfully submit that the amendments to the claims presented herein are sufficient to fully meet the requirements of 35 U.S.C. § 112, and respectfully solicit withdrawal of this rejection.

Prior to discussing the prior art rejections, Applicants wish to bring to the attention of the Examiner the grant of the corresponding European patent, the broadest claim of which is substantially similar to the claimed subject matter of claim 10. While Applicants' attorney recognizes that the grant of a patent in a foreign jurisdiction does not compel a grant in the U.S., the grant from the EPO is persuasive of patentability in the U.S., as the EPO, like the United States Patent and Trademark Office, performs a comprehensive search, and the laws of patentability with respect to novelty and obviousness ("lack of inventive step") are substantially similar in the EPO and the U.S.

The subject invention is directed to W/O and O/W emulsions of low relative viscosity, which are stable to coalescence and creaming, and which are prepared by use of a partly hydrophobic silica as a dispersing agent, and a special mixing process which allows for the emulsions to be free of coalescence and creaming. The term relative viscosity is defined in the claim, and is the ratio of the emulsion viscosity to the viscosity of the continuous phase (not the dispersed phase).

Emulsion stability of oil/water emulsions has been a longstanding problem, and has been successfully addressed in the past by careful choice of emulsifiers such as non-ionic surfactants. However, such surfactants are hydrophilic, and may have other undesirable properties depending upon the end use of the emulsion.

Partly water wettable (partly hydrophobic) silicas have been proposed to take the place of surfactants, but the emulsions prepared previously suffered from viscosity problems and stability problems. There are two stability problems associated with such emulsions. "Coalescence" (also called "precipitation" or "sedimentation") is the merging of smaller particles into larger ones, which separate as a separate continuous phase, as does the oil in a simple vinegar and oil Italian salad dressing. Creaming involves separation of the emulsion into distinct phases, both of which are still emulsions, but of different composition. The particle size may or may not increase during creaming.

Applicants have surprisingly and unexpectedly found that emulsions which are stable both to coalescence and creaming may be formed using partly hydrophobic silica as the dispersing medium, if all of the silica is first dispersed in a portion of the total continuous phase component to form a viscous ~~preemulsion~~, dispersion, the entire discontinuous (dispersed) phase is metered in with agitation, and finally, the remaining portion of the continuous phase is added. The emulsions thus prepared have a smaller mean particle size than emulsions prepared by other methods, and also a narrower particle size distribution. They also have a low viscosity relative to the viscosity of the continuous phase alone. Coalescence and creaming are eliminated or reduced to small amounts which are commercially tolerated.

Claims 10 - 12 and 15 - 18 have been rejected over copending and coassigned U.S. published application 2003/0175317 A1 ("*Barthel*"). Applicants respectfully traverse this rejection.

Barthel is also directed to the preparation of emulsions employing partly hydrophobic silica. Dr. Herbert Barthel is a coinventor of the claimed subject matter of the present application, and is well aware of the deficiencies of his prior patent, which is directed to the preparation of multiple emulsions of the O/W/O and W/O/W types. However, while emulsions prepared by the process of *Barthel* are storage stable with respect to coalescence, they neither have a desirably low viscosity nor are they stable to creaming. *Barthel* does not mention either of these properties, nor are they inherent, as shown by the comparative examples submitted herein, which will soon be submitted as a Rule 132 Declaration. The rejection under 35 U.S.C. § 102(b) should be removed for this reason.

Nor is the invention obvious over *Barthel*. *Barthel* does not teach or suggest the method of preparation of the claimed stable emulsions, and this method, as evidenced by the examples, produces a surprising and unexpected result. The claimed emulsions are prepared by a more complex process, involving more processing steps than taught by *Barthel*. The result is a considerably smaller particle size and narrower, almost Gaussian particle size

distribution. The relative viscosity is also low. These results are completely unpredictable in view of *Barthel*. Applicants have prepared direct, side-by-side comparative examples, as follows:

Example 1 according to US 20030175317 (Barthel et al.) (non-inventive):

A stainless steel beaker is charged with 56 g of an aqueous dispersion containing pyrogenic silica HKS® D (200 m²/g BET surface area; 71% residual silanol content) and 107 g of DI water resulting in a low viscosity dispersion. While stirring at 10000 rpm using an Ultraturrax® mixer, 150 g of polydimethylsiloxane (specific viscosity 100 mPas) was added slowly. After complete addition of polydimethylsiloxane the mixture was mixed for additional 5 minutes resulting in a white emulsion. Analytical data are summarized in Table 1.

Example 2 according to the invention:

A stainless steel beaker is charged with 56 g of an aqueous dispersion containing pyrogenic silica HKS® D (200 m²/g BET surface area; 71% residual silanol content) and 21 g of DI water in order to obtain a water content of 30 wt% in the preemulsion resulting in a highly viscous dispersion. While stirring at 10000 rpm using an Ultraturrax® mixer, 150 g of polydimethylsiloxane (specific viscosity 100 mPas) was added slowly. After complete addition of polydimethylsiloxane the mixture was mixed for additional 5 minutes. Finally, 86 g of DI water was added while stirring, resulting in a white emulsion. Analytical data are summarized in Table 1.

Table 1:

	Example 1	Example 2
Type	O/W	O/W
Φ_w/Φ_o	50/50	50/50
$d_{05} / \mu\text{m}$	1.7	1.2
$d_{50} / \mu\text{m}$	6.9	5.0
$d_{95} / \mu\text{m}$	30.0	9.2
$V_{\text{sep}} / \%$	30	< 1

The emulsion obtained from example 1 reveals a much broader droplet size distribution with a d_{95} of 30 μm compared to ca. 9 μm for the emulsion from example 2. Due to this broad droplet size distribution emulsion 1 shows strong phase separation with a depleted volume fraction V_{sep} of 30%. The phase separation (creaming) can be explained by the strong buoyancy of the large oil droplets. However, emulsion 2 shows no segregation.

In order to visualize the difference between emulsion 1 and emulsion 2 and therefore the advantage of the inventive process for making particle stabilized emulsions Figure 1 shows the results of droplet size measurements on emulsion 1 (= OG-131-A; red trace) versus emulsion 2 (= OG-131-B; blue trace). Again, a much broader droplet size distribution for emulsion 1 is seen.

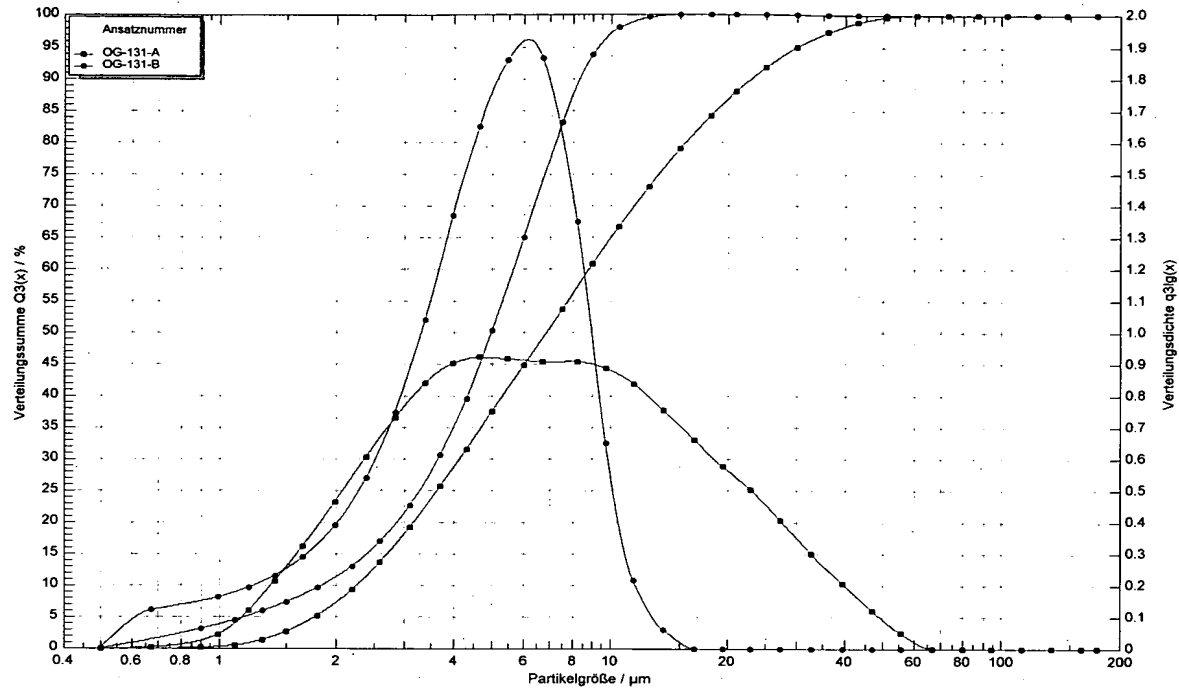


Figure 1: Droplet size distribution of emulsion 1 (= OG-131-A; red trace) emulsion 2 (= OG-131-B; blue trace) obtained by means of Fraunhofer laser light scattering.

This result is also supported by light microscopy (Figure 2) which clearly shows a broad droplet size distribution for emulsion 1 (Figure 2a) and a tight distribution for emulsion 2.

(Figure 2b).

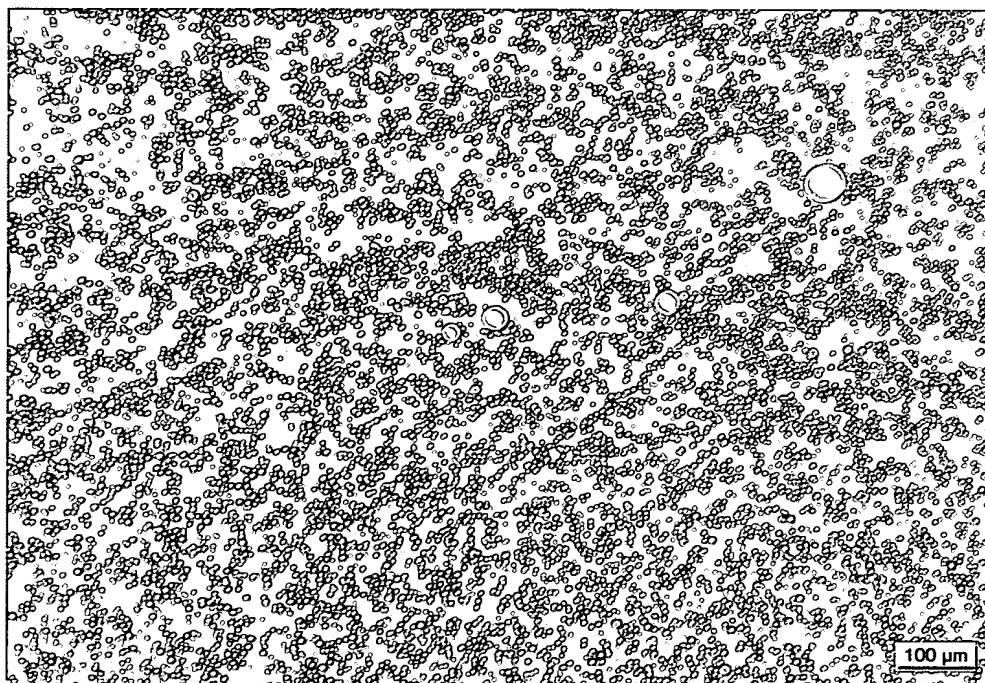
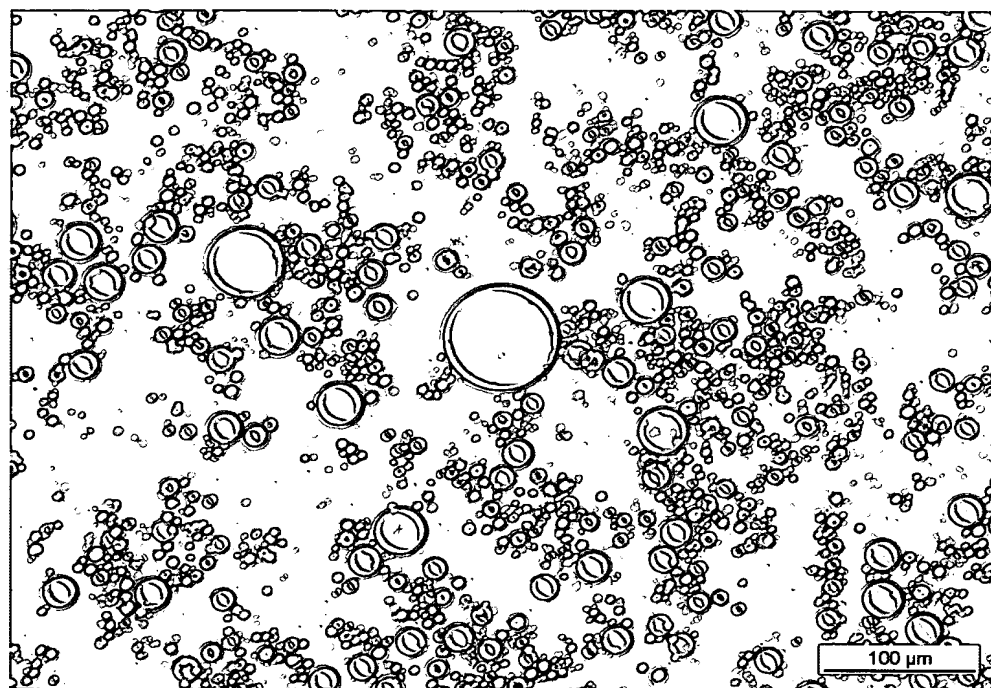


Figure 2: a) light microscopy of emulsion 1 and, b) light microscopy of emulsion 2.

The particle size distribution also strongly influences the intrinsic viscosity, which is the form factor used in the Krieger-Dougherty equation $\eta_{rel} = (1 - \Phi/0.74)^{([\eta] \cdot 0.74)}$ (see claim 10 of the present application). From theoretical calculations, an intrinsic viscosity $[\eta]$ of 2.5 is obtained for perfectly spherical and monodisperse particles. A broad particle size distribution results in an increase of $[\eta]$. Accordingly, the tight droplet size distribution of emulsion 2 results in an intrinsic viscosity of 5.7. For emulsion 1, which shows a broad droplet size distribution, an intrinsic viscosity of 23.4 is obtained.

The experiments clearly show the huge and surprisingly influence of the new process on emulsion properties. For commercial products, stability against phase separation is a must. This is only achieved by employing the inventive process according to the present patent application.

Withdrawal of the rejection of the claims under 35 U.S.C. § 103(a) over *Barthel* is respectfully solicited.

Claims 10 - 11 and 15 - 18 have been rejected under 35 U.S.C. § 102(b) and/or §103(a) over Binks et al. "Transitional Phase. . ." article published in Langmuir. ("*Binks I*"). As was the case with Dr. Barthel, Dr. Binks is also a coinventor, and is well aware of the deficiencies of the emulsions prepared in his scholarly paper.

The process taught by *Binks I* is the same as used by *Barthel*, and the same arguments apply. The emulsions of *Binks I* showed marked coalescence and creaming, even after only 30 minutes, as attested to by his figures. *Binks I* does not disclose, teach, or suggest a low viscosity emulsion stable against coalescence and creaming, nor any process for preparing such an emulsion. The wide range of particle sizes obtained by *Binks I* is evident from his photomicrographs. In many cases, bimodal particle size distributions were obtained.

The references to the background electrolyte concentration of *Binks I* are not understood, as first, none of Applicants' claims claim any electrolyte concentration, and second, the electrolyte concentration mentioned by *Binks* is the background electrolyte concentration, *i.e.* the electrolyte concentration used to calibrate his measuring instrument, not the electrolyte concentration of any emulsion. The reference to *Gottschalk-Gaudig* (also a coinventor) is not understood, and *Gottschalk-Gaudig* is not prior art, as the present application claims priority to German application 10 2004 014 704.3, filed on March 25, 2004, prior to the filing date of *Gottschalk-Gaudig* of October 21, 2004. Accompanying this response is an English translation of the German priority document which Applicants' attorney certifies is accurate.

Claims 10 – 13 and 15 – 18 have been rejected under 35 U.S.C. § 102(e) as anticipated and or obvious under 35 U.S.C. § 103(a) over *Gottschalk-Gaudig et al.* U.S. Patent No. 7,541,405 B2 ("*Gottschalk-Gaudig*"). Applicants respectfully traverse this rejection as *Gottschalk-Gaudig* is not a proper reference. Applicants however also do not agree with the grounds of rejection, as *Gottschalk-Gaudig* does not disclose the preparation method of the claimed dispersion, and the arguments relative to the "salt" content of the emulsions are not understood, as there is no limitation regarding this in any of the claims. Applicants also note that silica dispersing agents are not soluble. Silica is insoluble in both the aqueous phase and the oil phase. Withdrawal of the rejections over *Gottschalk-Gaudig* is respectfully solicited.

Claims 10 – 13 and 15 – 18 have been rejected under 35 U.S.C. § 102(a) or in the alternative under 35 U.S.C. § 103(a) over EP 1526153 A1 as evidenced by *Gottschalk-Gaudig*. EP 1 526 153 is not prior art, as it was first published on April 27, 2005, which is after both the present application's German priority date of March 24, 2004 and Applicants PCT filing date (designating the U.S.) of March 17, 2005. Thus, this rejection is respectfully traversed, and withdrawal of the rejections respectfully solicited.

Claims 10 – 12 and 15 – 18 have been rejected under 35 U.S.C. § 103(a) over *Binks I* in view of *Binks et al.*, “Effects of oil type. . .”, *Phys. Chem.* (“*Binks II*”). This rejection is not understood, particularly as formamide cannot be used to “replace” the aqueous component as it would then be a “non-aqueous” component. Neither *Binks I* nor *Binks II* disclose emulsions with low relative viscosity which are stable to coalescence and creaming, nor do either of *Binks I* or *Binks II* or their combination teach or suggest a method of preparing such low viscosity, stable emulsions. Withdrawal of the rejection of the claims over *Binks I* in view of *Binks II* is respectfully solicited.

Claims 10 – 13 and 15 – 18 have been rejected under 35 U.S.C. § 103(a) over *Barthel* U.S. Patent No. 5,686,054 (“*Barthel II*”). *Barthel II* is not relevant to the claimed invention. *Barthel* discloses highly hydrophobic, completely “apolar” silicas prepared by a special method of silylating which reacts all of the surface silanol groups. Note column 8, lines 62 – 66. No silanol groups are detectable, and the silica has no water-wettable fractions. It has a high thickening capacity in aqueous systems in which it is dispersible. These aqueous systems must also contain a significant fraction of a water soluble organic solvent, since the silica is completely non-dispersible otherwise. With methanol, for example, at least 50% of the aqueous phase must be methanol, since the methanol number is at least 50 and preferably higher. The methanol number is the percentage of methanol which must be present along with water to disperse this very highly hydrophobic silica. As is well known, such silica is completely impossible of forming a W/O or O/W emulsion as a stabilizer. Withdrawal of the rejection over *Barthel II* is respectfully solicited.

Claims 10 – 13 and 15 – 18 have been rejected for obviousness-type double patenting over *Gottschalk-Gaudig*. As indicated previously, *Gottschalk-Gaudig* is not prior art, nor does it disclose, teach, or suggest the claimed subject matter. However, as the filing of a terminal disclaimer will result in minimal loss of term of the patent, and in order to expedite prosecution, enclosed herewith is a Terminal Disclaimer over *Gottschalk-Gaudig*. Withdrawal of the obviousness-type double patenting rejection is respectfully solicited.

Claims 10 – 13 and 15 – 18 have been rejected for obviousness-type double patenting over claims 12 – 17 and 19 – 22 of copending application 11/573,379. Applicants do not acquiesce to this rejection as the present claims are clearly non-obvious over the '379 application. To expedite prosecution the terminal disclaimer includes the '379 application as well. Withdrawal of this rejection is thus respectfully solicited.

Applicants submit that the claims are now in condition for Allowance, and respectfully request a Notice to that effect. If the Examiner believes that further discussion will advance the prosecution of the Application, the Examiner is highly encouraged to telephone Applicants' attorney at the number given below.

The Petition fee of \$130.00 is enclosed. The Commissioner is hereby authorized to charge any additional fees or credit any overpayments as a result of the filing of this paper to Deposit Account No. 02-3978.

Respectfully submitted,

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